Hierarchy of relaxation times in the formation of an excitonic magnetic polaron in (CdMn)Te

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The circular polarization of the luminescence of excitonic magnetic polarons in $Cd_{1-x}Mn_xTe$, induced by an external magnetic field, was investigated experimentally and theoretically. It was determined that the lifetime of a magnetic polaron is longer than its formation time but shorter than the directional relaxation time of the polaron magnetic moment. © *1995 American Institute of Physics.*

The strong exchange interaction between localized charge carriers and magnetic ions in the crystal lattice in semimagnetic semiconductors leads to the formation of magnetic polaron states.[1] The formation of a localized excitonic magnetic polaron in an external magnetic field is due to three basic relaxation processes: 1) establishment and development of correlations between the carrier spins and the magnetic ions within the localization radius, 2) orientation of the polaron magnetic moment in an external magnetic field, and 3) recombination of the exciton. The ratios of the characteristic times of these processes determine the dynamics of the development of a magnetic polaron and the relation between its experimentally observed characteristics and the quasiequilibrium characteristics.

From the analysis of the luminescence of magnetic polaron states carried out in this study, it follows that the lifetime of a localized exciton falls between the characteristic formation time of a magnetic polaron and the directional relaxation time of its magnetic moment. In other words, over the lifetime of a localized exciton there is enough time for the magnetic moment of the ions that interact with exciton to relax to the equilibrium value of the magnetic moment of the polaron but with virtually no change in the direction of the moment.

We investigated $Cd_{1-x}Mn_xTe$ epitaxial layers, grown by the method of molecular-beam epitaxy on (100) CdTe substrates. The layers were 2 μm thick and the Mn-ion concentration x was varied from 0.15 to 0.35. The spin-relaxation of the photocarriers, which accompanies the formation of the excitonic magnetic polarons, was investigated by the method of polarized luminescence. The degree of circular polarization of the low-temperature ($T = 1.6$ K) photoluminescence, induced by an external magnetic field in the Faraday geometry, was measured with unpolarized continuous excitation in the spectrum of the free-exciton states. The investigated luminescence band corresponded to radiative

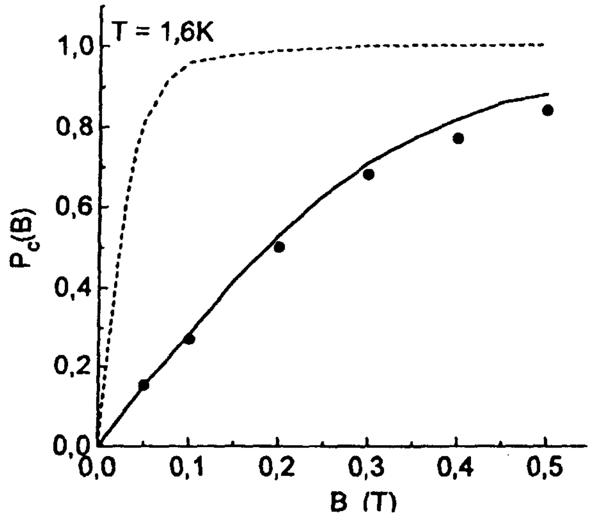

FIG. 1. Degree of circular polarization of luminescence induced by an external magnetic field in a $Cd_{0.76}Mn_{0.24}Te$ layer. Dots—Experiment; dashed line—calculation using Eq. (2) with $M_p = 120\mu_B$; solid line—calculation using Eq. (6) with the parameters given in the text.

recombination of localized excitonic magnetic polarons. The Zeeman splittings of the states of a free exciton, which characterize the exchange interaction between the carriers and the magnetic ions, were determined from the luminescence excitation spectra.[2] The formation of excitonic magnetic polarons in these structures was investigated in detail by the method of time-resolved spectroscopy.[3] In the $Cd_{0.76}Mn_{0.24}Te$ layer, for which we present the experimental results in the present paper, the energy of a magnetic polaron is $E_p = 23$ meV and it remains virtually constant in the experimental interval of magnetic fields up to 0.5 T. The formation time of a magnetic polaron is equal to 90 ps, which is much shorter than its recombination time (350 ps). A magnetic polaron thus reaches its equilibrium energy within its lifetime.

To determine the ratio of the lifetime and directional relaxation time of the magnetic moment ($\mathbf{M}_p$) of a polaron, we measured the degree of circular polarization $P_c(B)$ of the luminescence as a function of the external magnetic field; the measurements are represented by dots in Fig. 1. The theoretical calculation of this dependence for the equilibrium distribution of $\mathbf{M}_p$ was performed using the following formula derived in Ref. 4:

$$P_{c,mp}(b_1) = \frac{\exp(b_1)(b_1^2 - b_1) + \exp(-b_1)(b_1^2 + b_1)}{\exp(b_1)(b_1^2 - b_1 + 1) - \exp(-b_1)(b_1^2 + b_1 + 1)}, \tag{1}$$

where $b_1 = M_p B/(k_B T) = B/B_1$ is the external magnetic field measured in units of $B_1 = (k_B T)/M_p$, k_B is Boltzmann's constant, and T is the temperature. Good agreement between the experimental data and the computational results can be obtained by using the magnetic moment as an adjustable parameter, as was done in Ref. 4. However, the value obtained for M_p in this manner is eight times smaller than the value computed below directly from the experimentally measured spin splittings of the free-hole states.[1)]

If the spin splitting $E_z(B)$ of the states of a free hole with projections $\pm 3/2$ onto the external magnetic field is determined, then the magnetic moment of the polaron and the characteristic field B_1 are estimated as

$$M_p=\frac{E_p}{B_p}\approx\frac{1}{2}\left(\frac{dE_z}{dB}\right)_{B=0}, \quad B_1=\frac{k_BTB_p}{E_p}\approx\frac{2k_BT}{\left(\frac{dE_z}{dB}\right)_{B=0}}. \tag{2}$$

Here E_p is the exchange energy of a magnetic polaron, and B_p is the average exchange field generated by a localized hole at magnetic ions located within the polaron. The magnitude of this field is determined from the condition $E_p=E_z(B_p)/2\approx B_p dE_z/2dB$, where the last approximate equality is valid since $E_z(B)$ is linear.

The Zeeman splitting of the excitonic states in $Cd_{1-x}Mn_xTe$ is described by a modified Brillouin function with the effective temperature T_0 and effective spin S_{eff}.[2] For the experimental sample with $x=0.24$, these parameters were $T_0=8.4$ K and $xS_{\text{eff}}=0.1$. The linear approximation describes satisfactorily the function $E_z(B)$ up to the values $B=B_p\approx 3.2$ T. These experimentally determined values of the parameters give for the magnetic moment of the polaron $M_p=120\mu_B$, where μ_B is the Bohr magneton. The *equilibrium* circular polarization of the localized magnetic polarons as a function of the external magnetic field constructed for each value of the magnetic moment using Eq. (1) is represented by the dashed line in Fig. 1. We see that it gives a much higher value of P_c than is observed experimentally. It is obvious that the polarization of the polarons is of a *nonequilibrium* character.

We now consider the second limiting case, in which the directional equilibration time of the polaron magnetic moments is much longer than the polaron lifetime. Following Refs. 5 and 6, we consider a model in which the polaron magnetic moment is oriented in the same direction as the total magnetic moment of its constituent ions at the moment of carrier localization; i.e., we assume that at the moment of exciton localization the carrier spins are oriented along the direction of the total magnetic moment of the Mn ions within the localization length. The polaron magnetic moment will then increase in magnitude over the lifetime of an exciton, and in the process the exchange energy of the magnetic polaron increases, but there is not enough time for the initial direction of the magnetic moment to change.

To calculate theoretically the polarization of the luminescence in this model, it is necessary to know the directional distribution function of the magnetic moment for magnetization fluctuations which transform into magnetic polarons after localization of the exciton. In the experiments described above, because the applied external fields are weak, the polarization of the magnetic ions is far from being saturated, so that a Gaussian approximation of the distribution function of the magnetic moment can be used to describe the fluctuations of the magnetic moment of the ions that interact with the hole (Ref. 7; see also Refs. 1, 5, and 6):

$$\varphi(\mathbf{m},\mathbf{b})=\frac{1}{\pi^{3/2}}\exp\{-|\mathbf{m}-\mathbf{b}_2|^2\}; \quad \mathbf{b}_2=\frac{\mathbf{B}}{B_2}, \tag{3}$$

where $B_2 = 2kT/\sqrt{2\langle M^2\rangle}$, $\mathbf{m} = \mathbf{M}/\sqrt{2\langle M^2\rangle}$ is the fluctuation magnetic moment in units of $\sqrt{2\langle M^2\rangle}$, and $\langle M^2\rangle$ is the average value of the squared projection of the magnetic moment of a fluctuation onto an arbitrary direction.

The distribution (3) makes it possible to easily obtain the well-known[7] relation between $\langle M^2\rangle$ and the magnetic susceptibility, from which we find the following expression on the basis of the linear approximation for $M(B)$:

$$\langle M^2\rangle = k_B T \frac{d\langle M\rangle}{dB} = \frac{k_B T}{2B_p}\frac{dE_z}{dB} \approx \frac{k_B T}{E_p}\left(\frac{dE_z}{2dB}\right)^2. \tag{4}$$

Substituting this expression into Eq. (3), we obtain the final expression for the parameter B_2, which relates the external magnetic field B with the dimensionless field b_2:

$$B_2 = 2\frac{\sqrt{2k_B T E_p}}{dE_z/dB} = B_1\sqrt{\frac{2E_p}{k_B T}} = \sqrt{\frac{2kT}{E_p}}B_p. \tag{5}$$

All quantities appearing in this equation can be measured directly. This makes it possible to use the distribution (3) to calculate directly the circular polarization of the luminescence, determined by the equilibrium distribution of the directions of the fluctuation magnetic moment, as a function of the magnetic field:

$$P_{c,f}(b) = \frac{\int\int\int d^3m(I_+(\gamma) - I_-(\gamma))\varphi(\mathbf{m},\mathbf{b})}{\int\int\int d^3m(I_+(\gamma) + I_-(\gamma))\varphi(\mathbf{m},\mathbf{b})}, \tag{6}$$

where γ is the angle between the fluctuation magnetic moment $\mathbf{m}$ and the direction of the magnetic field $\mathbf{b}$, and $I_\pm \propto (1 \pm \cos\gamma)^2$ is the probability for the emission of right-hand and left-hand circularly polarized photons accompanying the recombination of a magnetic polaron whose magnetic moment makes an angle γ with the direction of observation.

The solid curve in Fig. 1 was constructed according to Eq. (6) using the following experimentally determined values of the parameters: $dE_z/dB = 14.4$ meV/T, $E_p = 23$ meV, and $T = 1.6$ K. One can see that it describes well the experimental dependence $P_c(B)$. On the basis of this model we were able to obtain good agreement with the experimental data in the entire experimental range of compositions from $x = 0.15$ to $x = 0.35$. This allows us to conclude that the fluctuation model adequately describes the real situation in the test crystals.

We shall now discuss the reason why the rapid relaxation of the fluctuation magnetic moment to the equilibrium polaron magnetic moment is not accompanied by rapid relaxation of the directional distribution of the magnetic moments of the magnetic polarons. To explain this difference, we call attention to the fact that the spins of the constituent magnetic ions of a polaron are subjected to the sum of the external magnetic field $\mathbf{B}$ and the exchange field $\mathbf{B}_p$, whose direction is the same as that of $\mathbf{M}$. Let us assume that the relaxation of the total magnetic moment of all ions in the polaron is described by the very simple equation

$$\frac{d\mathbf{m}}{dt} = -\frac{1}{T_1}\left[\mathbf{m} - \left(\mathbf{b} + b_p\frac{\mathbf{m}}{m}\right)\right], \tag{7}$$

where $b = B/B_2$ and $b_p = B_p/B_2$. This equation contains the only relaxation time (T_1). We take into account here the fact that the external field b is $\ll b_p$. The rate of change of the magnetic moment, which determines the value of the magnetic polaron energy and the rate of change of the direction of the magnetic polaron, is then given by the equations

$$\left(\frac{\mathbf{m}}{m}\frac{d\mathbf{m}}{dt}\right) = -\frac{1}{T_1}[m - b\cdot\cos\vartheta - b_p] \approx -\frac{1}{T_1}[m - b_p], \tag{8}$$

$$\frac{d\cos\vartheta}{dt} = \frac{\mathbf{b}}{b}\frac{d}{dt}\left(\frac{\mathbf{m}}{m}\right) = -\frac{1}{T_1}\frac{b}{m}(\cos\vartheta - 1), \tag{9}$$

where ϑ is the angle between the magnetic moment and the external magnetic field. At the outset when $m \approx 1$, $d\cos\vartheta/dt$ is of the order of $b/T_1 < 1/T_1 < 10^{10}\ \mathrm{s}^{-1}$. However, the system exists at such values of the magnetic moment only for a short time, of the order of T_1/b_p, so that there is virtually no time for the angle ϑ to change ($\Delta\vartheta \approx b/b_p \ll 1$). Next, the magnetic moment is of the order of b_p and the directional equilibration rate decreases by a factor of b_p. An estimate of the magnitude of the dimensionless polaron field with the parameters employed above gives $b_p/b = B_p/B \geq 15$. The estimate obtained in this manner for the characteristic directional relaxation time of the polaron magnetic moments (2×10^{-9} s) is almost five times greater than the polaron lifetime (3.5×10^{-10} s). This explains the experimentally observed hierarchy of the characteristic times of the relaxational formation processes of magnetic polarons.

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[1)]A hole is singled out because holes interact four times more strongly with magnetic ions than do electrons.

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